

CLAIMS

560
1. An ester selected from the group consisting of
alicyclic or aromatic dicarboxylic acid diesters

5 represented by the formula (E)



wherein A represents a cyclohexane ring, a cyclohexene ring
or a benzene ring, X is a hydrogen atom or methyl, R^x and R^y
are the same or different and each is a branched-chain

10 alkyl group having 3 to 18 carbon atoms, a straight-chain
alkyl group having 1 to 18 carbon atoms, a straight-chain
alkenyl group having 2 to 18 carbon atoms or a cycloalkyl
group having 3 to 10 carbon atoms, provided that when A is
a benzene ring, R^x and R^y are different from each other and
15 the group -COOR^x and the group -COOR^y are attached to
adjacent two carbon atoms of the benzene ring, the ester
having the following properties:

- 1) a total acid number of 0.05 mgKOH/g or less,
- 2) a sulfated ash content of 10 ppm or less,
- 20 3) a sulfur content of 20 ppm or less,
- 4) a phosphorus content of 20 ppm or less,

- 5) a peroxide value of 1.0 meq/kg or less,
6) a carbonyl value of 10 or less,
7) a volume resistivity of $1 \times 10^{11} \Omega \cdot \text{cm}$ or more,
8) a hydroxyl value of 3 mgKOH/g or less, and
5 9) a water content of 100 ppm or less.

2. An ester selected from the group consisting of
(I) alicyclic dicarboxylic acid diesters represented
by the formula (1)



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wherein A^1 represents a cyclohexane ring or cyclohexene ring,
X is a hydrogen atom or methyl, R^1 and R^2 are the same or
different and each is a branched-chain alkyl group having 3
to 18 carbon atoms, a straight-chain alkyl group having 1
15 to 18 carbon atoms, a straight-chain alkenyl group having 2
to 18 carbon atoms or a cycloalkyl group having 3 to 10
carbon atoms; and

(II) alicyclic or aromatic adjacent dicarboxylic acid
mixed diesters represented by the formula (4)



wherein A represents a cyclohexane ring, a cyclohexene ring
or a benzene ring, X is a hydrogen atom or methyl, R⁵ and R⁶
5 are different from each other and each is a branched-chain
alkyl group having 3 to 18 carbon atoms, a straight-chain
alkyl group having 1 to 18 carbon atoms, a straight-chain
alkenyl group having 2 to 18 carbon atoms or a cycloalkyl
group having 3 to 10 carbon atoms, and the group -COOR⁵ and
10 the group -COOR⁶ are attached to two adjacent carbon atoms
of the cyclohexane ring, cyclohexene ring or benzene ring
represented by A, the ester having the following
properties:

- 1) a total acid number of 0.05 mgKOH/g or less,
- 15 2) a sulfated ash content of 10 ppm or less,
- 3) a sulfur content of 20 ppm or less,
- 4) a phosphorus content of 20 ppm or less,
- 5) a peroxide value of 1.0 meq/kg or less,
- 6) a carbonyl value of 10 or less,
- 20 7) a volume resistivity of $1 \times 10^{11} \Omega \cdot \text{cm}$ or more,
- 8) a hydroxyl value of 3 mgKOH/g or less, and

9) a water content of 100 ppm or less.

3. An alicyclic dicarboxylic acid diester represented by the formula (1)

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wherein A¹ represents a cyclohexane ring or cyclohexene ring, X is a hydrogen atom or methyl, R¹ and R² are the same or different and each is a branched-chain alkyl group having 3 to 18 carbon atoms, a straight-chain alkyl group having 1 to 18 carbon atoms, a straight-chain alkenyl group having 2 to 18 carbon atoms or a cycloalkyl group having 3 to 10 carbon atoms; the alicyclic dicarboxylic acid diester having the following properties:

- 15 1) a total acid number of 0.05 mgKOH/g or less,
 2) a sulfated ash content of 10 ppm or less,
 3) a sulfur content of 20 ppm or less,
 4) a phosphorus content of 20 ppm or less,
 5) a peroxide value of 1.0 meq/kg or less,
20 6) a carbonyl value of 10 or less,
 7) a volume resistivity of 1 x 10¹¹Ω·cm or more,

- 8) a hydroxyl value of 3 mgKOH/g or less, and
- 9) a water content of 100 ppm or less.

4. The alicyclic dicarboxylic acid diester according
5 to claim 3 wherein A¹ is a cyclohexane ring and X is a
hydrogen atom, or A¹ is a cyclohexene ring and X is a
hydrogen atom, or A¹ is a cyclohexene ring and X is methyl,
and the two ester groups -COOR¹ and -COOR² are attached to
1- and 2-positions of the cyclohexane ring or cyclohexene
10 ring represented by A¹.

5. The alicyclic dicarboxylic acid diester according
to claim 4 wherein R¹ and R² are the same and each
represents straight-chain or branched-chain alkyl group
15 having 3 to 11 carbon atoms, A¹ is a cyclohexane ring or
cyclohexene ring and X is a hydrogen atom.

6. A process for preparing an alicyclic dicarboxylic
acid diester represented by the formula (1)



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wherein A¹ represents a cyclohexane ring or cyclohexene ring,

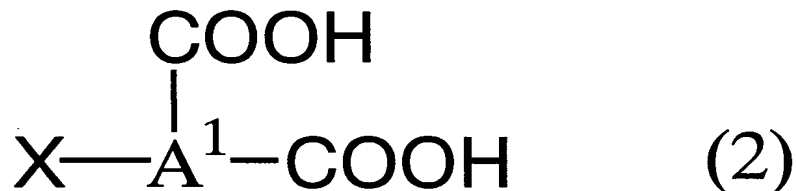
X is a hydrogen atom or methyl, R¹ and R² are the same or different and each is a branched-chain alkyl group having 3 to 18 carbon atoms, a straight-chain alkyl group having 1 to 18 carbon atoms, a straight-chain alkenyl group having 2 to 18 carbon atoms or a cycloalkyl group having 3 to 10 carbon atoms; and having the following properties:

- 1) a total acid number of 0.05 mgKOH/g or less,
- 2) a sulfated ash content of 10 ppm or less,
- 3) a sulfur content of 20 ppm or less,
- 10 4) a phosphorus content of 20 ppm or less,
- 5) a peroxide value of 1.0 meq/kg or less,
- 6) a carbonyl value of 10 or less,
- 7) a volume resistivity of $1 \times 10^{11} \Omega \cdot \text{cm}$ or more,
- 8) a hydroxyl value of 3 mgKOH/g or less, and
- 15 9) a water content of 100 ppm or less,

the process comprising the steps of

(i) subjecting

a) an alicyclic dicarboxylic acid represented by the formula (2)



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wherein A¹ and X are as defined above, or an anhydride thereof, and

b) an aliphatic monohydric alcohol having 1 to 18 carbon atoms or an alicyclic monohydric alcohol having 3 to 10 carbon atoms each having a peroxide value of 1.0 meq/kg or less

5 to esterification reaction in the absence of a catalyst or in the presence of a sulfur-free and phosphorus-free catalyst, or subjecting

a') an alicyclic dicarboxylic acid diester represented by the formula (3)



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wherein A¹ and X are as defined above, R³ and R⁴ are the same or different and each is a branched-chain alkyl group having 3 or 4 carbon atoms or a straight-chain alkyl group having 1 to 4 carbon atoms, and

15 b') an aliphatic monohydric alcohol of 5 to 18 carbon atoms or an alicyclic monohydric alcohol of 3 to 10 carbon atoms each having a peroxide value of 1.0 meq/kg or less to ester interchange reaction in the absence of a catalyst or in the presence of a sulfur-free and phosphorus-free
20 catalyst,

to thereby obtain a reaction mixture containing the diester

represented by the formula (1),

(ii) removing excess starting materials from the reaction mixture obtained in step (i) to thereby obtain the diester in a crude form,

5 (iii) neutralizing the crude diester obtained in step (ii) and washing the neutralized crude diester with water,

(iv) purifying the crude diester neutralized and washed with water in step (iii) by treatment with 1 to 4 kinds of adsorbents, and

10 (v) dehydrating the diester purified in step (iv).

7. The process for preparing the alicyclic dicarboxylic acid diester according to claim 6 wherein said alcohols under b) or b') used in step (i) has a carbonyl value of 15
15 or less.

8. The process for preparing the alicyclic dicarboxylic acid diester according to claim 6 wherein the esterification reaction or the ester interchange reaction
20 in step (i) is carried out in an inert gas atmosphere or in an inert gas stream.

9. The process for preparing the alicyclic dicarboxylic acid diester according to claim 6 wherein the
25 esterification reaction or the ester interchange reaction

in step (i) is carried out in the presence of a sulfur-free and phosphorus-free catalyst, the catalyst being selected from the group consisting of tetra(C₃-C₈ alkyl) titanate, titanium oxide, titanium hydroxide, sodium alkoxide of 1 to 4 carbon atoms, sodium hydroxide, C₃-C₁₂ fatty acid tin salt, tin oxide, tin hydroxide, zinc oxide, zinc hydroxide, lead oxide, lead hydroxide, aluminum oxide and aluminum hydroxide.

10 10. The process for preparing the alicyclic dicarboxylic acid diester according to claim 6 wherein in step (iii), the neutralization is carried out until the crude diester has a total acid number of 0.05 mgKOH/g or less after being washed with water, and the crude diester
15 is washed with water until the pH of the washings used for the washing becomes neutral.

 11. The process for preparing the alicyclic dicarboxylic acid diester according to claim 6 wherein the
20 treatment with adsorbents in step (iv) is carried out using 2 to 4 kinds of the adsorbents selected from the group consisting of activated carbon, activated alumina, silica gel, silica-alumina, activated clay, zeolite, magnesia, calcia, diatomaceous earth, hydrotalcite, ion exchange
25 resins of the non-sulfonic acid type and synthetic

hydrotalcite.

12. A refrigerator lubricating oil comprising the alicyclic dicarboxylic acid diester according to any one of
5 claims 3-5.

13. A refrigerator lubricating oil comprising the alicyclic dicarboxylic acid diester obtainable by the process according to any one of claims 6-11.
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14. An alicyclic or aromatic adjacent dicarboxylic acid mixed diester represented by the formula (4)



wherein A represents a cyclohexane ring, a cyclohexene ring
15 or a benzene ring, X is a hydrogen atom or methyl, R⁵ and R⁶
are different from each other and each is a branched-chain
alkyl group having 3 to 18 carbon atoms, a straight-chain
alkyl group having 1 to 18 carbon atoms, a straight-chain
alkenyl group having 2 to 18 carbon atoms or a cycloalkyl
20 group having 3 to 10 carbon atoms, and the group -COOR⁵ and
the group -COOR⁶ are attached to two adjacent carbon atoms

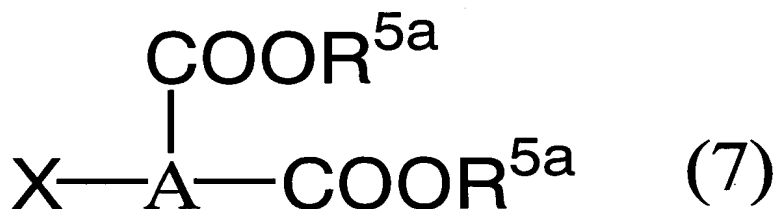
of the cyclohexane ring, cyclohexene ring or benzene ring represented by A; and having the following properties:

- 1) a total acid number of 0.05 mgKOH/g or less,
- 2) a sulfated ash content of 10 ppm or less,
- 5 3) a sulfur content of 20 ppm or less,
- 4) a phosphorus content of 20 ppm or less,
- 5) a peroxide value of 1.0 meq/kg or less,
- 6) a carbonyl value of 10 or less,
- 7) a volume resistivity of $1 \times 10^{11} \Omega \cdot \text{cm}$ or more,
- 10 8) a hydroxyl value of 3 mgKOH/g or less, and
- 9) a water content of 100 ppm or less.

15 15. An alicyclic or aromatic adjacent dicarboxylic acid mixed diester according to claim 14 wherein A is a cyclohexane ring or cyclohexene ring, X is a hydrogen atom, R⁵ is a straight-chain alkyl group having 1 to 5 carbon atoms or a branched-chain alkyl group having 3 to 5 carbon atoms, and R⁶ is a straight-chain or branched chain alkyl group having 6 to 11 carbon atoms, and when A is a
20 cyclohexene ring, the group -COOR⁵ and group -COOR⁶ are present at the 1- and 2-positions and the double bond is present between the 4- and 5-positions.

25 16. An ester mixture of
(1) an alicyclic or aromatic adjacent dicarboxylic

acid di(lower alkyl) ester represented by the formula (7)



5 wherein A represents a cyclohexane ring, a cyclohexene ring
or a benzene ring, X is a hydrogen atom or methyl and R^{5a} is
a branched-chain alkyl group having 3 to 5 carbon atoms, a
straight-chain alkyl group having 1 to 5 carbon atoms, a
straight-chain alkenyl group having 2 to 5 carbon atoms or
10 a cycloalkyl group having 3 to 5 carbon atoms, and the two
-COOR^{5a} groups are the same and attached to two adjacent
carbon atoms of the cyclohexane ring, cyclohexene ring or
benzene ring represented by A;

(2) an alicyclic or aromatic adjacent dicarboxylic
15 acid mixed diester represented by the formula (4a)



wherein A and X are as defined in the formula (7), and R^{5a} and R^{6a} are different from each other and R^{5a} is as defined in the formula (7), and R^{6a} is a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and the group -COOR^{5a} and the group -COOR^{6a} are attached to two adjacent carbon atoms of the cyclohexane ring, cyclohexene ring or benzene ring represented by A, and

(3) an alicyclic or aromatic adjacent dicarboxylic acid di(higher alkyl)ester represented by the formula (8)



wherein A, X and R^{6a} are as defined in the formula (4a), and the two -COOR^{6a} groups are the same and attached to two adjacent carbon atoms of the cyclohexane ring, cyclohexene ring or benzene ring represented by A,

the ester mixture having the following properties:

- 1) a total acid number of 0.05 mgKOH/g or less,
- 2) a sulfated ash content of 10 ppm or less,

- 3) a sulfur content of 20 ppm or less,
- 4) a phosphorus content of 20 ppm or less,
- 5) a peroxide value of 1.0 meq/kg or less,
- 6) a carbonyl value of 10 or less,
- 5 7) a volume resistivity of $1 \times 10^{11} \Omega \cdot \text{cm}$ or more,
- 8) a hydroxyl value of 3 mgKOH/g or less, and
- 9) a water content of 100 ppm or less.

17. The ester mixture according to claim 16 wherein
10 the alicyclic or aromatic adjacent dicarboxylic acid mixed
diester represented by the formula (4a) under (2) is
present in a proportion of 100, the alicyclic or aromatic
adjacent dicarboxylic acid di(lower alkyl) ester
represented by the formula (7) under (1) is present in a
15 proportion of 5-300, and the alicyclic or aromatic adjacent
dicarboxylic acid di(higher alkyl) ester under (3) is
present in a proportion of 7-500, wherein the proportions
are expressed in terms of area ratio as determined from a
gas chromatogram of the ester mixture.

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18. The ester mixture according to claim 16 wherein the
ester mixture is a mixture of an alicyclic adjacent
dicarboxylic acid di(lower alkyl) ester represented by the
formula (7), an alicyclic adjacent dicarboxylic acid mixed
25 diester represented by the formula (4a) and an alicyclic

adjacent dicarboxylic acid di(higher alkyl) ester represented by the formula (8), the ester mixture having a trans isomer/cis isomer ratio of 0/100 to 80/20 (by area % as determined by gas chromatography).

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19. A process for preparing an alicyclic or aromatic adjacent dicarboxylic acid mixed diester or an ester mixture, the alicyclic or aromatic adjacent dicarboxylic acid mixed diester being represented by the formula (4)



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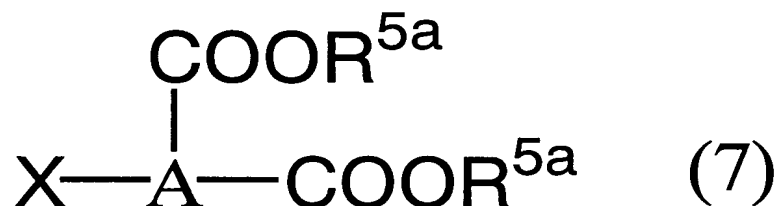
wherein A represents a cyclohexane ring, a cyclohexene ring or a benzene ring, X is a hydrogen atom or methyl, R⁵ and R⁶ are different from each other and each is a branched-chain alkyl group having 3 to 18 carbon atoms, a straight-chain alkyl group having 1 to 18 carbon atoms, a straight-chain alkenyl group having 2 to 18 carbon atoms or a cycloalkyl group having 3 to 10 carbon atoms (particularly, R⁵ is a straight-chain alkyl group having 1 to 5 carbon atoms or a branched-chain alkyl group having 3 to 5 carbon atoms, R⁶ is a straight-chain or branched-chain alkyl group having 6 to 11 carbon atoms), and the group -COOR⁵ and the group -COOR⁶

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are attached to two adjacent carbon atoms of the cyclohexane ring, cyclohexene ring or benzene ring represented by A; and said ester mixture being a mixture of

- (1) an alicyclic or aromatic adjacent dicarboxylic
5 acid di(lower alkyl) ester represented by the formula (7)



- wherein A and X are as defined in the formula (4), and R^{5a} represents a branched-chain alkyl group having 3 to 5
10 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and the two -COOR^{5a} groups are the same and attached to two adjacent carbon atoms of the cyclohexane ring,
15 cyclohexene ring or benzene ring represented by A,

- (2) an alicyclic or aromatic adjacent dicarboxylic acid mixed diester represented by the formula (4a)



wherein A and X are as defined in the formula (7), and R^{5a} and R^{6a} are different from each other and R^{5a} is as defined in the formula (7), and R^{6a} is a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and the group -COOR^{5a} and the group -COOR^{6a} are attached to two adjacent carbon atoms of the cyclohexane ring, cyclohexene ring or benzene ring represented by A, and

(3) an alicyclic or aromatic adjacent dicarboxylic acid di(higher alkyl)ester represented by the formula (8)



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wherein A, X and R^{6a} are as defined in the formula (4a), and the two -COOR^{6a} groups are the same and attached to two adjacent carbon atoms of the cyclohexane ring, cyclohexene ring or benzene ring represented by A, and the alicyclic or aromatic adjacent dicarboxylic acid mixed

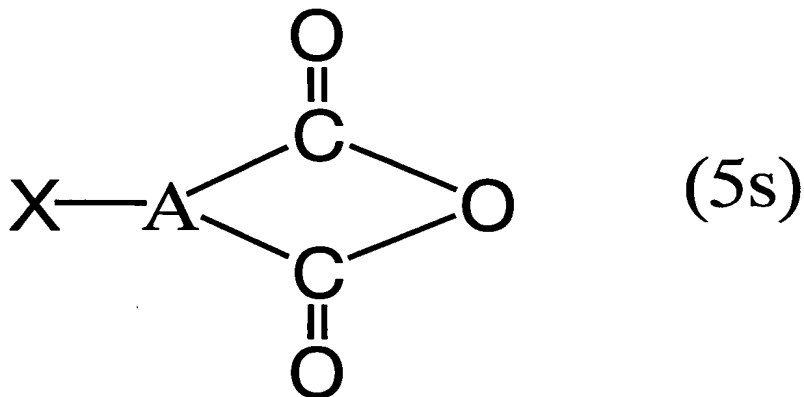
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diester or the ester mixture having the following properties:

- 1) a total acid number of 0.05 mgKOH/g or less,
- 2) a sulfated ash content of 10 ppm or less,
- 5 3) a sulfur content of 20 ppm or less,
- 4) a phosphorus content of 20 ppm or less,
- 5) a peroxide value of 1.0 meq/kg or less,
- 6) a carbonyl value of 10 or less,
- 7) a volume resistivity of $1 \times 10^{11} \Omega \cdot \text{cm}$ or more,
- 10 8) a hydroxyl value of 3 mgKOH/g or less, and
- 9) a water content of 100 ppm or less,

the process comprising the steps of

- (i) (a) subjecting an alicyclic or aromatic adjacent dicarboxylic anhydride represented by the formula
- 15 (5s)



wherein A and X are as defined above and "alcohol

component 1" (namely, a single alcohol or alcohol mixture comprising a monohydric alcohol having 1 to 5 carbon atoms (P) and a monohydric alcohol having 6 to 18 carbon atoms (Q) wherein (P):(Q) is 0.1:99.9 to 100:0 (molar ratio)) to esterification reaction to thereby give an alicyclic or aromatic adjacent dicarboxylic acid monoester represented by the formula (5)



wherein A, X and R⁵ are as defined above, and the group -COOR⁵ and the group -COOH are attached to two adjacent carbon atoms of the cyclohexane ring, cyclohexene ring or benzene ring represented by A,

(b) subjecting the alicyclic or aromatic adjacent dicarboxylic acid monoester represented by the formula (5) obtained in step (a) and "alcohol component 2" (namely, a single alcohol or alcohol mixture comprising a monohydric alcohol having 1 to 5 carbon atoms (S) and a monohydric alcohol having 6 to 18 carbon atoms (T) wherein (S):(T) is 0:100 to 99.9:0.1 (molar ratio)) to a further esterification

reaction in the absence of a catalyst or in the presence of a sulfur-free and phosphorus-free catalyst

- 5 to thereby give a reaction mixture containing said ester mixture of (1) the alicyclic or aromatic adjacent dicarboxylic acid di(lower alkyl) ester represented by the formula (7), (2) the alicyclic or aromatic adjacent dicarboxylic acid mixed diester represented by the formula (4a), and (3) the
- 10 alicyclic or aromatic adjacent dicarboxylic acid di(higher alkyl) ester represented by the formula (8),
- (ii) removing excess starting materials from the reaction mixture obtained in step (i) to thereby obtain the
- 15 ester mixture in a crude form,
- (iii) neutralizing the crude ester mixture obtained in step (ii) and washing the neutralized crude ester mixture with water,
- (iv) purifying the crude ester mixture neutralized and
- 20 washed with water in step (iii) by treatment with 1 to 4 kinds of adsorbents,
- (v) dehydrating the diester purified in step (iv) to thereby give the ester mixture having the properties 1) to 9), and if desired,
- 25 (vi) separating the alicyclic or aromatic adjacent

dicarboxylic acid mixed diester mixture represented by the formula (4a) under (2) to thereby give the alicyclic or aromatic adjacent dicarboxylic acid mixed diester represented by the formula (4).

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20. The process according to claim 19 wherein the ester mixture contains the alicyclic or aromatic adjacent dicarboxylic acid mixed diester represented by the formula (4a) under (2) in a proportion of 100, the alicyclic or aromatic adjacent dicarboxylic acid di(lower alkyl) ester represented by the formula (7) under (1) in a proportion of 5-300, and the alicyclic or aromatic adjacent dicarboxylic acid di(higher alkyl) ester under (3) in a proportion of 7-500, wherein the proportions are expressed in terms of area ratio as determined from a gas chromatogram of the ester mixture.

21. The process according to claim 19 wherein the ester mixture is a mixture of an alicyclic adjacent dicarboxylic acid di(lower alkyl) ester represented by the formula (7), an alicyclic adjacent dicarboxylic acid mixed diester represented by the formula (4a) and an alicyclic adjacent dicarboxylic acid di(higher alkyl) ester represented by the formula (8), the ester mixture having a trans isomer/cis isomer ratio of 0/100 to 80/20 (by area %

as determined by gas chromatography).

22. The process according to claim 19 wherein the monohydric alcohol of 1 to 5 carbon atoms (P) constituting
5 said "alcohol component 1" is an alcohol composed of a branched-chain alkyl group having 3 to 5 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and a
10 hydroxyl group, and the monohydric alcohol of 6 to 18 carbon atoms (Q) is an alcohol composed of a branched-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl
15 group having 6 to 10 carbon atoms, and a hydroxyl group, and
wherein the monohydric alcohol of 1 to 5 carbon atoms (S) constituting said "alcohol component 2" is an alcohol composed of a branched-chain alkyl group having 3 to 5
20 carbon atoms, a straight-chain alkyl group having 1 to 5 carbon atoms, a straight-chain alkenyl group having 2 to 5 carbon atoms or a cycloalkyl group having 3 to 5 carbon atoms, and a hydroxyl group, and the monohydric alcohol of 6 to 18 carbon atoms (T) is an alcohol composed of a
25 branched-chain alkyl group having 6 to 18 carbon atoms, a

straight-chain alkyl group having 6 to 18 carbon atoms, a straight-chain alkenyl group having 6 to 18 carbon atoms or a cycloalkyl group having 6 to 10 carbon atoms, and a hydroxyl group.

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23. The process according to claim 19 wherein said "alcohol component 1" is a monohydric alcohol of 1 to 5 carbon atoms and said "alcohol component 2" is a monohydric alcohol of 6 to 18 carbon atoms.

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24. The process according to claim 19 wherein said "alcohol component 1" and said "alcohol component 2" have a peroxide value of 1.0 meq/kg or less.

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25. The process according to claim 24 wherein said "alcohol component 1" and said "alcohol component 2" further have a carbonyl value of 15 or less.

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26. The process according to claim 19 wherein said esterification reactions in steps (a) and (b) of step (i) are carried out in an inert gas atmosphere or in an inert gas stream.

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27. The process according to claim 19 wherein said reaction in step (a) of step (i) is carried out in the

absence of a catalyst and said esterification reaction in step (b) is carried out in the presence of a catalyst selected from the group consisting of tetra(C₃-C₈ alkyl) titanate, titanium oxide, titanium hydroxide, sodium alkoxide of 1 to 4 carbon atoms, sodium hydroxide, C₃-C₁₂ fatty acid tin salt, tin oxide, tin hydroxide, zinc oxide, zinc hydroxide, lead oxide, lead hydroxide, aluminum oxide and aluminum hydroxide.

10 28. The process according to claim 19 wherein the proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is 10 to 90 mole%, relative to the total amount [(P)+(Q)+(S)+(T)] of alcohol component 1 [(P)+(Q)] used in the first stage esterification reaction and alcohol
15 component 2 [(S)+(T)] used in the second-stage esterification reaction, and

- 1) the whole amount of the monohydric alcohol of 1 to 5 carbon atoms is used as (P) in the first-stage esterification reaction and 0 mole% of said monohydric
20 alcohol is used in the second-stage esterification reaction, when said proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is not less than 10 mole% and not greater than 50 mole%, and
- 2) the monohydric alcohol of 1 to 5 carbon atoms is used
25 as (P) in an amount of 50 mole% relative to the total

amount [(P)+(Q)+(S)+(T)] in the first-stage esterification reaction and the rest of said monohydric alcohol of 1 to 5 carbon atoms is used as (S) in the second-stage esterification reaction, when said
5 proportion of the monohydric alcohol of 1 to 5 carbon atoms [(P)+(S)] is more than 50 mole% and not more than 90 mole% relative to the total amount [(P)+(Q)+(S)+(T)].

29. The process according to claim 19 wherein in step
10 (iii), the neutralization is carried out until the total acid number of the crude ester mixture becomes 0.05 mgKOH/g or less after being washed with water, and the crude ester mixture is washed with water until the pH of the washings used for the washing becomes neutral.

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30. The process according to claim 19 wherein the treatment with adsorbents in step (iv) is carried out using 2 to 4 kinds of the adsorbents selected from the group consisting of activated carbon, activated alumina, silica
20 gel, silica-alumina, activated clay, zeolite, magnesia, calcia, diatomaceous earth, hydrotalcite, ion exchange resins of the non-sulfonic acid type and synthetic hydrotalcite.

25 ^{SUB} 31. A refrigerator lubricating oil comprising the

~~alicyclic or aromatic adjacent dicarboxylic acid mixed diester according to any one of claims 14 and 15 or the ester mixture according to any one of claims 16-18.~~

- 5 32. A refrigerator lubricating oil comprising the alicyclic or aromatic adjacent dicarboxylic acid mixed diester or the ester mixture obtainable by the process according to any one of claims 19-30.

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